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Airborne reduced nitrogen: ammonia emissions from agriculture and other sources

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Abstract

Ammonia is a basic gas and one of the most abundant nitrogen-containing compounds in the atmosphere. When emitted, ammonia reacts with oxides of nitrogen and sulfur to form particles, typically in the fine particle size range. Roughly half of the PM_{2.5} mass in eastern United States is ammonium sulfate, according to the US EPA. Results from recent studies of PM2.5 show that these fine particles are typically deposited deep in the lungs and may lead to increased morbidity and/or mortality. Also, these particles are in the size range that will degrade visibility. Ammonia emission inventories are usually constructed by multiplying an activity level by an experimentally determined emission factor for each source category. Typical sources of ammonia include livestock, fertilizer, soils, forest fires and slash burning, industry, vehicles, the oceans, humans, pets, wild animals, and waste disposal and recycling activities. Livestock is the largest source category in the United States, with waste from livestock responsible for about 3×10^9 kg of ammonia in 1995. Volatilization of ammonia from livestock waste is dependent on many parameters, and thus emission factors are difficult to predict. Despite a seasonal variation in these values, the emission factors for general livestock categories are usually annually averaged in current inventories. Activity levels for livestock are from the USDA Census of Agriculture, which does not give information about animal raising practices such as housing types and grazing times, waste handling systems, and approximate animal slurry spreading times or methods. Ammonia emissions in the United States in 1995 from sources other than livestock are much lower; for example, annual emissions are roughly 8×10^8 kg from fertilizer, 7×10^7 kg from industry, 5×10^7 kg from vehicles and 1×10^8 kg from humans. There is considerable uncertainty in the emissions from soil and vegetation, although this category may also be significant. Recommendations for future directions in ammonia research include designing experiments to improve emission factors and their resolution in all significant source categories, developing mass balance models, and refining of the livestock activity level data by eliciting judgment from experts in this field. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Ammonia; Emission inventory; Livestock; Animal waste; Fertilizer; Emission factors; Ammonium nitrate; Ammonium sulfate

1. Introduction

Ammonia is the most prevalent basic gas in the atmosphere, and therefore it plays a major role in the neutralization of precipitation, cloudwater and aerosols (Aneja et al., 2000a,b). Deposition of ammonia and ammonium contributes to water and soil acidification and may cause forest damage (Bouwman and van der Hoek, 1997; Bouwman et al., 1997; Lee and Dollard, 1994). Also, increased nitrogen supply to terrestrial and aquatic ecosystems can cause eutrophication (Walker et al., 2000). Ammonia gas has a relatively short lifetime in the atmosphere of a few hours to a few days (Warneck, 1988; Dentener and Crutzen, 1994). In contrast, the ammonium ion, as an aerosol, may have a

lifetime on the order of 1-15 days (Aneja et al., 1998). Gaseous ammonia typically reacts with oxides of nitrogen and sulfur to form ammonium sulfate and ammonium nitrate particles, as shown in reactions (1) and (2) (Seinfeld and Pandis, 1998).

$$NH_3(g) + HNO_3(g) \Longrightarrow NH_4NO_3(g)$$
 (1)

$$2NH_3(g) + H_2SO_4(g) = (NH_4)_2SO_4(s)$$
 (2)

Ammonia comprises a significant portion of the PM_{2.5} mass; in the eastern United States, 47% of the PM_{2.5} mass is ammonium sulfate according to an extensive set of monitoring data (EPA, 1995) (Fig. 1).

Results from recent studies of PM_{2.5} show that these fine particles can deposit deep in the lungs, which may lead to increased morbidity and/or mortality (EPA, 1996). Also,

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these particles are in the size range that will degrade visibility (Seinfeld and Pandis, 1998). With impending changes in particulate matter air quality standards, states must develop or adjust their existing state implementation plans (SIPs) to demonstrate compliance with the standards. In order to compose acceptable SIPs, computer models of atmospheric chemistry are being improved to predict concentrations of several pollutants. These models use meteorological data and emission inventories of chemical species as input. Since ammonia is such a significant portion of particulate mass, an accurate emission inventory is necessary for input to air quality models to predict concentrations.

To estimate the total emission rate of a compound within a region (e.g., in mass/time), an emission factor, or mass of the compound emitted per unit of activity, is multiplied by an activity level. Typically, activity levels are expressed as volume of fuel burned/time, mass of material produced/time, kilometers traveled/time, or other dimensions that define the size of a source. An inventory may thus be composed of emission rates for each type of source in a region. Because ammonia is not federally regulated in the United States, there have been only a few attempts to compile emission inventories for this compound. Results of an effort by E. H.Pechan and Associates (Roe et al., 1998) are shown in Fig. 2. More than half of the emissions in the United States are from livestock, estimated using emission factors (mass of ammonia/time emitted from one animal) multiplied by activity level (number of animals).

A more recent inventory prepared by Carnegie Mellon University (CMU) shows that some sources may contribute more ammonia than previously thought (Strader et al., 2001). For example, emissions calculated for the United States in 1995 for the most important categories include: 3.4×10^9 kg from livestock, 7.7×10^8 kg from fertilizer application, 1.5×10^8 kg from domestic animals, 1.3×10^8 kg from wild animals, 1.1×10^8 kg from humans, 7.0×10^7 kg from industry, 4.7×10^7 kg from mobile sources, and 6.9×10^4 kg from publicly owned treatment works (POTWs). There may also be significant emissions from soil but values are highly uncertain. In this paper, we

Composition of Fine Particles (PM_{2.5}) in Eastern U.S. (EPA 1995)

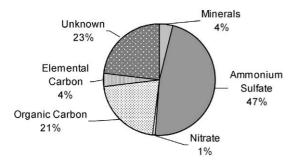


Fig. 1. Fine particle composition (EPA, 1995).

Ammonia Emission Inventory: National Particulates Inventory (E.H. Pechan & Associates)

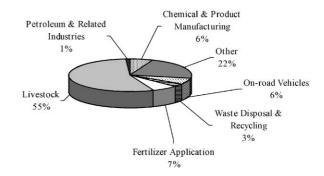


Fig. 2. Ammonia emission inventory for the United States in 1990 (after Roe et al., 1998).

compare and discuss published emission factors and activity levels for ammonia in the United States. We focus on livestock as the most important category but also consider fertilizer, soil, and several minor categories.

2. Ammonia emissions from livestock

Nitrogen in livestock food sources that does not end up in a product (e.g., milk or eggs) or that does not get absorbed by the body, is excreted by the animal. Nitrogen excreted in animal feces is typically bound up in organic compounds; limited data suggest that only 1-5% of this nitrogen volatilizes as ammonia (Lockyer and Whitehead, 1990). However, some studies have shown steady yearround emissions from stored slurry that may be due to slow release of ammonia from the feces (Patni and Jui, 1991). Nitrogen in the urine is in the form of urea, CO(NH₂)₂, which can rapidly hydrolyze to form ammonium carbonate. As shown in reactions (3), (4) and (5), decomposition of ammonium carbonate frees up ammonium ions that can volatilize as gaseous ammonia (Jarvis and Pain, 1990). Hydrolysis is facilitated by the enzyme urease, which is abundant in soils and plant roots as well as in animal feces (Elzing and Monteny, 1997; Whitehead, 1990).

$$CO(NH_2)_2 + 3H_2O \rightarrow (NH_4)_2CO_3 + H_2O$$
 (3)

$$(NH_4)_2CO_3 + H_2O \rightarrow 2NH_4^+ + HCO_3^- + OH^-$$
 (4)

$$NH_4^+ \to NH_3 + H^+ \tag{5}$$

The amount of ammonia that volatilizes depends on factors such as the amount of nitrogen in the food source, size and species of the animal, housing conditions of the animal, humidity, temperature, and animal waste handling practices.

In the past few decades, in the United States, there has been a trend toward specialized livestock farms. It is more

Table 1 Major factors influencing ammonia emissions from livestock

	Stable	Storage	Spreading	Grazing
Waste characteristics	Dry matter content, pH, N/NH ₄ ⁺ content, the presence of bedding	Dry matter content, pH, N/NH ₄ ⁺ content, formation of a surface crust	Dry matter content, pH, N/NH ₄ ⁺ content, formation of a surface crust	Dry matter content, pH, N/NH ₄ ⁺ content, formation of a surface crust
Environmental conditions	Air temperature, wind speed/ ventilation rate, relative humidity	Air temperature, wind speed/ ventilation rate, relative humidity, rainfall	Air temperature, wind speed, relative humidity, rainfall, soil temperature and characteristics	Air temperature, wind speed, relative humidity, rainfall, soil temperature and characteristics
Waste management variations	Time before waste removal, type of stable	Type of storage, loading rate of waste, frequency of emptying storage	Time of day, application rate, application method	Length of time and time of day animals are grazing

profitable to focus on producing one type of animal rather than a mixture of animals and crops. Since these large production facilities have limited land space, animals are more often confined in buildings rather than being allowed to graze. For example, pigs and poultry are almost always confined in buildings. Beef cattle may graze in pasture but are later confined in feedlots for several months for fattening prior to slaughter. Dairies are currently making the transition to confined operations; some of the smaller dairy farms are still grazing their cows.

With increasing production, specialization, and confinement disposal of animal wastes is now problematic. In the past, most wastes were spread on fields and used as fertilizer for the crops. Now, however, animal production facilities may not have enough land to incorporate all of the waste as fertilizer, and it may not be economical to transport it to locations needing fertilizer (Lander et al., 1998). Excess spreading of animal wastes could result in nitrogen-rich runoff or groundwater contamination (Lander et al., 1998; Kohn, 1998). To alleviate these problems, many large farms now have management systems for storing the manure so that spreading can be done at appropriate times.

Emission factors have been reported in the literature for different phases of the life cycle of animal wastes, such as on the floor of the barn, in a storage container, or applied to the land as fertilizer. Ammonia emissions from waste generated during confinement and subsequent storage and spreading are nearly an order of magnitude higher than emissions from waste while the animal is grazing in pasture (Roe et al., 1998). This is due to the soil taking up much of the nitrogen. If the animal is grazing for its entire life, there is only one phase to be measured. Currently inventories aggregate the emission factors from each phase into one emission factor, the amount of ammonia emitted per animal per year.

One of the more widely used compilations of ammonia emission factors reported in the literature was put together by Asman (1992), focusing on data for the Netherlands. The review includes cattle, hogs, chickens, horses, and turkeys with emission factors broken down further into age or weight categories. An ammonia emission inventory for the United States was developed using emission factors from the Asman report by Battye et al. (1994). However, the animal categories used by Asman are slightly different than those used by Battye et al. For example, the Asman report does not give an emission factor for the general population of beef cows. For this animal category, the Battye et al report used the Asman emission factor for dairy cows. But dairy cows will excrete more nitrogen than beef cows, given their more nitrogen-rich diet (Acker and Cunningham, 1998). Besides this problem, use of a composite emission factor for all phases of the life cycle of animal waste causes inaccuracies. A better representation of emissions could be obtained by multiplying an emission factor for each phase by a typical amount of time that the waste spends in that phase. The animal category for these emission factors could then be as narrow as the activity data allow.

One problem with the use of published emission factors is that they are typically annual averages, and there is considerable variability in the reported values. Since emission factors depend on temperature, relative humidity, and winds one would expect seasonal variations. In addition, animal raising practices change throughout the year as animals grow, are transported, and slaughtered. An annually averaged emission factor does not capture these changes. Furthermore, many of the emission factors have been developed in Europe, where climate and animal raising practices differ from those in the United States. Studies that have attempted to determine ammonia emissions as a function of such parameters as air temperature, relative humidity, pH of the waste, and dry matter content of the waste show large variability due to the numerous parameters that affect volatilization (Tables 1 and 2).

Table 2 Popular waste management types

Stable	Storage	Spreading			
Storage below slatted floors, manure pack, alley- scrape with subsequent storage, alley-scrape with no storage, barn floors flushed with water, milking parlor, feedlot	Top or bottom loaded, covered or uncovered, above- or under-ground tank, earthen pond or lagoon, storage under slatted floors, oxidation ditch, compost pile	Injection, slurry spreading, spreading as farm yard manure, incorporation after spreading			

Table 3

Average ammonia emission factors for dairy cattle with the standard deviation enclosed in parenthesis

Phase of emission	Number of values	Emission factor
Stable plus storage	5	15.5(11.4) kg NH ₃ /cow-year
Stable	5	9.3(3.2) kg NH ₃ /cow-year
Storage	3	5.4(2.5) kg NH ₃ /cow-year
Storage in above- ground storage structure	12	24(16.3)% of total N lost as NH ₃
Storage in earthen pond or lagoon	9	37.7(20.1)% of total N lost as NH ₃
Storage below slatted floors	2	45(7.1)% of total N lost as NH ₃
Storage as manure pack	10	22.3(15.5)% of total N lost as NH ₃
Daily scrape and haul (no storage)	5	22(13)% of total N lost as NH ₃
Spreading	8	13.2(4.5) kg NH ₃ /cow-year
Grazing	11	6.4 (5.1) kg NH ₃ /cow-year
-	11	10.5(6.2)% of excreted N lost as NH ₃

The reference lists at the end of this paper represent various emission factor studies from the literature for the stable, storage, and spreading phases of wastes from dairy cattle, pigs and poultry. Grazing is included for dairy cattle. These animal categories have been shown to be the largest livestock sources of ammonia in previous inventories. Average emission factors for dairy cattle from these references are shown in Table 3.

2.1. Influence of parameters on emission factors

Most studies of factors influencing emissions from animal wastes have focused on spreading of dairy cattle slurry.

One might expect the volatilization of ammonia to be strongly influenced by air temperature; higher temperatures should be associated with greater volatilization rates. However, other factors may override the influence of temperature. For example, at high temperatures and dry conditions, a surface crust may form on the slurry that inhibits volatilization. Also, at cold temperatures, frozen soil may prevent infiltration of the slurry, resulting in greater long-term release of ammonia to the atmosphere. Fig. 3 shows volatilization of ammonia from spreading of dairy cattle slurry as a function of temperature, expressed as the percent of total ammonical nitrogen released. Volatilization increases as temperature increases, but there is much scatter in the data. The study done by Menzi et al. (1998) captures the largest range of temperatures and has the highest correlation, with an R^2 value of 0.66. The two points from this dataset that are circled are the result of special circumstances. The circled point at 14.5 °C falls slightly lower than the regression line, probably because a visible surface crust on the slurry was inhibiting ammonia emissions. The circled point at 2.9 °C falls above the regression line because, although the loss rate was low, it remained steady over a long period of time and therefore cumulative losses were high. This was most likely due to frozen ground that inhibited infiltration of the slurry into the soil.

The dry matter content of animal waste slurry depends on the amount of dilution with water. One would expect more dilute slurries to have lower cumulative volatilization, as the ammonical nitrogen is less concentrated. Fig. 4 shows the percent of total ammonical nitrogen volatilized from spreading dairy cattle slurry as a function of dry matter content. Volatilization increases as the dry matter content of the slurry increases. The study by Sommer and Olesen (1991) does not show a correlation between ammonia loss and temperature (Fig. 3), but does show a correlation between

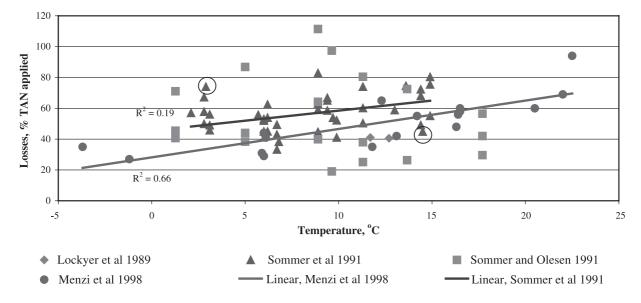


Fig. 3. Volatilization of ammonia expressed as a percent of total ammoniacal nitrogen versus air temperature (Lockyer et al., 1989; Menzi et al., 1998; Sommer and Olesen, 1991; Sommer et al., 1991). The two circled data points of Menzi et al. reflect special conditions (see text).

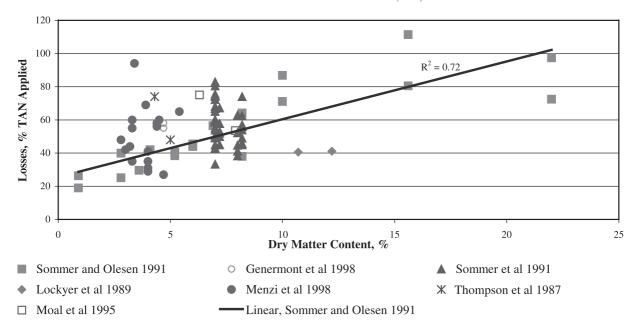


Fig. 4. The volatilization of ammonia expressed as a percent of total ammonical nitrogen versus dry matter content of the slurry (Genermont et al., 1998; Lockyer et al., 1989; Menzi et al., 1998; Moal et al., 1995; Sommer et al., 1991; Sommer and Olesen, 1991; Thompson et al., 1987).

ammonia loss and dry matter content. The other studies do not capture a sufficient range of dry matter content to see a correlation, considering scatter in the data.

Air temperature and dry matter content are just two of the parameters influencing ammonia emissions from spread slurry; many other parameters also affect emissions. For example, the presence of bedding may have a significant impact on volatilization, as bedding may absorb the urine and prevent it from hydrolyzing (Jacobson Larry, 2000). This makes it difficult to choose an emission factor for spread slurry.

2.2. Activity levels

The most complete inventory of animals in the United States, the Census of Agriculture, is prepared by the National Agricultural Statistics Service (NASS) of the US Department of Agriculture (USDA). The Census is administered once every 5 years, and the inventories reported are for December 31 of the inventory year. By law, anyone receiving a census is required to complete it, even if he does not qualify as having operated a farm. A farm is defined as "any place from which \$1000 or more of agricultural products were produced and sold, or normally would have been sold, during the census year" (USDA, 1997). Statistical analyses are performed on the census data to account for unreported inventories and to quantify uncertainty. Census results have proven to be relatively reliable and are widely used. Even though results are obtained for each individual farming operation, they are published only at the national, state, and county level to avoid disclosing data for individual farms. Therefore, data at the county level represent the best spatial resolution available for the ammonia

emission inventory from livestock. The categories of animals reported by the census are shown in Table 4. Animal inventories for these categories are reported for total farms as well as for farms with the herd size denominations shown in the table.

Some of the categories do not allow for accurate application of existing emission factors. For example, the 'steers, steer calves, bulls, and bull calves' category includes types of animals with very different emission factors; emissions from bulls are about 28 kg NH₃/bull-year and emissions from calves are about 5 kg NH₃/calf-year (Asman, 1992). Sales information is also given in the census; the number of sales of 'cattle fattened on grain and concentrates' is given but not the total population of cattle in this category. Also, the census does not include any information on animal raising practices or waste management systems. Since the literature reports emission factors for each phase of animal waste life cycle for the most popular animal raising practices and structures, activity levels for these divisions are needed to put this information to use. For dairy cattle, the census does not report types of housing, types and duration of manure storage, approximate manure spreading times, or separate numbers of animals that are confined and that are grazing.

2.3. Mass balance approach

Recently, several studies reported in the literature have used a mass balance approach on the animal wastes, based on distinctions between housing types, manure storage types, and grazing and spreading times, to estimate ammonia emissions from dairy cattle (Hutchings et al., 1996; Pollet et al., 1998; Ferguson et al., 2001). According to the

Table 4
Livestock categories reported by the USDA Census of Agriculture

Cattle and calves	Hogs and pigs	Poultry
Total cattle and calves	Total hogs and pigs	Total poultry
Cows and heifers that had calved, total	Hogs and pigs used or to be used for breeding	Layers and pullets 13 weeks old and older
Cows and heifers that had calved, beef cows	Other hogs and pigs	Layers 20 weeks old and older
Cows and heifers that had calved, milk cows	Litters of pigs farrowed between Dec. 1 of	Pullets 13 weeks old and older but less than
	preceding year and Nov. 30	20 weeks old
		Pullet chicks and pullets less than 13 weeks old
Heifers and heifer calves	Litters of pigs farrowed between Dec. 1 of preceding year and May 31	Broilers and other meat-type chickens
		Turkeys, total
Steers, steer calves, bulls, and bull calves	Litters of pigs farrowed between June 1 and Nov. 30	Turkey hens kept for breeding
		Ducks, geese and other poultry
Herd Size Denominations (head of animals)		
1-9; 10-19; 20-49; 50-99; 100-199;	1-24; 25-99; 100-199; 200-499; 500-999;	1-49; 50-99; 100-399; 400-3199; 3200-
200-499; 500 and more	1000 and more	9999; 10,000-19,999; 20,000-49,999;
		50,000-99,999; 100,000 and more

mass balance equation, the total ammonia emitted from one dairy cow over a particular length of time is equal to the sum of emissions from the animal's waste. The sum includes the time when the waste is in the barn, while it is in storage, while it is being field-applied, and from waste deposited while the animal is grazing during that time. The fraction of excreted nitrogen that is volatilized as ammonia, in each of these phases of the life cycle of the waste, can be obtained from the literature. The fraction is shown either as the mass of ammonia emitted per phase or as the percent of the nitrogen in the waste in each phase that volatilizes as ammonia. However, further resolution of the activity levels would be necessary to use this information for ammonia emission inventory purposes. The surveying of farmers or elicitation of agricultural experts' opinions may be necessary to obtain estimates of animal populations that are handled with different types of management practices. When these life cycle-partitioned activity levels are combined with ammonia emission factors from the literature, more accurate ammonia emissions can be estimated.

3. Ammonia emissions from fertilizer application

Fertilizer application is typically considered the second or third most important source of ammonia on a national level, depending on whether the inventory includes soil emissions. Existing inventories usually estimate the contribution from fertilizer application to be 10–20% of the national total (Roe et al., 1998; Strader et al., 2001). Activity level data for fertilizer application can be obtained from the Association of American Plant and Food Control Officials (AAPFCO) that reports fertilizer sales to farmers at the county level. The AAPFCO data sets contain county-level resolution for the top producing states for each crop (usually about 30 states), and state-level resolution for the

other states. Although there are almost 200 different commercial fertilizers used in the United States, roughly 13 of them account for the majority of fertilizer use. The total use of the remaining fertilizers is less than 3–4%. Emission factors for the 13 major fertilizers can be obtained from Battye et al. (1994) and Asman (1992).

Ammonia emissions from fertilizer application have a strong temporal component that has previously been ignored; yearly averages have been used in most inventories. Significantly more fertilizer is applied in the spring and fall than in the summer and winter, corresponding to crop cycles. Accounting for the timing of fertilizer application allows monthly resolution in emissions, included in the CMU inventory (Davidson, 2002).

4. Ammonia emissions from soil

Emission from soil is the most uncertain source category in an ammonia emission inventory, but soil has the potential to be a major source. A 1990 inventory for the San Joaquin Valley in California estimated soil emissions to be 40% of the total (Sonoma Technology, 1998). High quality emission factors for soil types are not available, and the physics of ammonia-surface exchange is not well understood. A soil—plant canopy system can be a source of ammonia emissions under certain conditions and a sink under other conditions (Milford et al., 2000). Because of this uncertainty, many existing inventories simply do not include emissions from soil despite its possible importance.

Cass et al. (1982) applied Anderson land use codes as activity levels for soil emissions. This is possibly the best existing method for estimating soil emissions on a national level. The emission factors reported by Cass et al. are very uncertain annual averages, but better data are not currently available.

5. Ammonia emissions from minor sources

5.1. Mobile sources

Although mobile sources are important for other airborne contaminants, they are minor sources of ammonia on a national scale, typically comprising only a few percent of the total. To estimate mobile source emissions in ammonia inventories, data have been obtained from state transportation departments that give vehicle miles traveled per year for each county in the nation. Corresponding emission factors have been obtained from Battye et al. (1994).

5.2. Industry

Industry also plays a small role in ammonia emissions, comprising only a few percent of the national total. The EPA's Toxic Release Inventory (TRI) database (EPA, 1995) reports approximate ammonia emissions for industry directly, so it is not necessary to use the activity level/emission factor model employed for other source categories.

5.3. Publicly owned treatment works

Ammonia emissions from wastewater treatment plants are not included in the TRI. Activity levels for all publicly owned treatment works (POTWs) in the nation can be obtained from the Office of Water of the US Environmental Protection Agency (1996) and emission factors can be obtained from Battye et al. (1994).

5.4. Humans

Ammonia emissions from human breath and perspiration comprise a few percent of the national ammonia emission inventory. The US Census Bureau (1990, 1997) reports population data that can represent activity levels for this category, and emission factors can be obtained from Battye et al. (1994).

5.5. Domestic animals

Activity levels for animals kept as pets can be obtained from the American Veterinary Medical Association (1997). Corresponding emission factors can be obtained from Battye et al. (1994).

5.6. Wild animals

Several categories of wild animals have been included in existing ammonia emission inventories. The CMU inventory includes three: bear, deer, and elk. Activity levels can be obtained from the American Bear Association (1993), the Quality Deer Management Association (2000), and the Rocky Mountain Elk Foundation (1995), and emission factors are available from Botsford et al. (1997).

5.7. Forest fires and slash burning

To compute ammonia emissions from forest fires and slash burning, the number of acres burned in an area can be multiplied by typical fuel loading for that region, and then by an emission factor. For the CMU inventory, data for the number of acres burned per state were obtained from the National Interagency Fire Center (1994). Typical fuel loading amounts were obtained from EPA (1998), and an emission factor was calculated by combining an emission factor for carbon monoxide from forest fires (EPA, 1998) and a ratio of ammonia to carbon monoxide concentrations measured in plumes from forest fires (Hegg et al., 1988).

6. Summary and conclusions

Ammonia is a known precursor to atmospheric aerosols. Since the accuracy of the predictions of particulate matter concentrations from an air quality model is only as good as the quality of the input data, there is a need for high quality emission inventories of important atmospheric compounds, including ammonia. There is much room for improvement in the current ammonia emission inventories, in resolution as well as the accuracy of the data. Livestock waste is the largest source of atmospheric ammonia in the United States, so decreasing the uncertainty in livestock waste emissions would have a major impact on the accuracy of the entire ammonia emission inventory.

The livestock ammonia emission inventory could be improved by using better emission factors that become available as more experiments are done, using activity levels that incorporate animal raising practices, and by improving spatial and temporal resolution. Further experimentation with ammonia emissions from each phase of animal waste's life cycle could establish relationships between emissions and such parameters as temperature, relative humidity, wind speed, rainfall, nitrogen content, dry matter content, and pH of the waste. Further experimentation with emissions from waste storage is especially needed, as most available data are outdated. Besides the need for more empirical data, a better understanding of the chemistry involving ammonia emissions from livestock waste is desirable. This will permit modeling of ammonia emissions for many different situations and environmental conditions. Better data on the life cycle of animal wastes in the stable, in storage facilities, and on land could be used in a mass balance modeling approach to calculate total ammonia emissions with improved temporal resolution and county-level spatial resolution.

Information on dates of fertilizer application has permitted monthly resolution of emissions from this source category. In contrast, emissions from soil are still poorly understood. Major improvements are needed, both in our understanding of ammonia-surface exchange in different soil—plant canopy systems and in the development of emission factors for specific systems. Other minor source

categories, such as mobile sources, industry, POTWs, humans, domestic animals, wild animals, forest fires, and slash burning will also require further experimentation as the resolution for the ammonia emission inventories continues to be improved.

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References

- Acker D, Cunningham M, editors. Animal science and industry. New Jersey: Prentice Hall; 1998. p. 45–75.
- American Bear Association. http://www.americanbear.org/. Fax giving bear populations for 1993.
- American Veterinary Medical Association. http://www.avma.org/. US Pet Ownership and Demographics Sourcebook; 1997.
- Aneja VP, Chauhan JP, Walker JT. Characterization of atmospheric ammonia emissions from swine waste storage and treatment lagoons. J Geophys Res (Atmospheres) 2000a;105:11535-45.
- Aneja VP, Roelle PA, Murray GC, Southerland J, Erisman JW, Fowler D, et al. Atmospheric nitrogen compounds II: emissions, transport, transformation, deposition and assessment. Atmos Environ 2000b;35: 1903–11.
- Asman WAH, Ammonia emissions in Europe: Updated emission and emission variations. Report No 229471008, National Institute of Public Health and Environmental Protection, Bilthoven, The Netherlands; May 1992. p. 1–45.
- Battye R, Battye W, Overcash C, Fudge S. Development and Selection of Ammonia Emission Factors. Report No. 68-D3-0034, Atmospheric Research and Exposure Assessment Laboratory, US Environmental Protection, Research Triangle Park, NC. p. 2-1 to 12, 3-8, 5-4, 6-1, 6-2.
- Botsford CW, Chitjian M, Koizumi J, Wang Y, Gardner L, Winegar E. Gridded Ammonia Emission Inventory Update for the South Coast Air Basin. Diamond Bar, CA: South Coast Air Quality Management District; 1997.
- Bouwman AF, Van Der Hoek KW. Scenarios of animal waste production and fertilizer use and associated ammonia emission for the developing countries. Atmos Environ 1997;31:4095–102.
- Bouwman AF, Lee DS, Asman WAH, Dentener FJ, Van Der Hoek KW, Oliver JGJ. A global high-resolution emission inventory for ammonia. Glob Biogeochem Cycles 1997;11:561–87.
- Cass GR, Gharib S, Peterson M, Tilden JW. The origin of ammonia emissions to the atmosphere in an urban area. Report 82-6, Environmental Quality Laboratory, California Institute of Technology, Pasadena, CA; 1982
- Davidson CI. Department of Civil and Environmental Engineering, Carnegie Mellon University, personal communication; 2002.

- Dentener FJ, Crutzen PJ. A three-dimensional model of the global ammonia cycle. J Atmos Chem 1994;19:331–69.
- Elzing A, Monteny GJ. Ammonia emission in a scale model of a dairy-cow house. Trans Am Soc Agric Eng 1997;40:713–20.
- Ferguson J, Dou Z, Ramberg C. An assessment of ammonia emissions from dairy facilities in Pennsylvania. Optimizing nitrogen management in food and energy production and environmental protection. Proc. 2nd International Nitrogen Conference on Science and Policy. The Scientific World, vol. 1. 2001. p. 348–55.
- Genermont S, Cellier P, Flura D, Morvan T, Laville P. Measuring ammonia fluxes after slurry spreading under actual field conditions. Atmos Environ 1998;32:279–84.
- Hegg DA, Radke LF, Hobbs PV. Ammonia emissions from biomass burning. Geophy Res Lett 1988;15:335–7.
- Hutchings NJ, Sommer SG, Jarvis SC. A model of ammonia volatilization from a grazing livestock farm. Atmos Environ 1996;30:589-99.
- Jacobson Larry D. Department of Biosystems and Agricultural Engineering, University of Minnesota, personal communication; 2000 Nov. 22.
- Jarvis SC, Pain BF. Ammonia volatilization from agricultural land. Proc No 298. London: The Fertilizer Society; 1990. p. 5–33.
- Kohn RA. Utilization by cattle of the nitrogen in forage crops. ACS Symp Series, Nitrogen-Containing Macromolecules in the Bio-and Geosphere, 214th National Meeting. Las Vegas, NV: American Chemical Society; 1998. p. 278–92.
- Lander CH, Moffitt D, Alt K, Nutrients available from livestock manure relative to crop growth requirements. Resource Assessment and Strategic Planning Working Paper 98-1, http://www.nhq.nrcs.usda.gov/land/ pubs/nlweb.html, Natural Resources Conservation Service, US Department of Agriculture, February 1998.
- Lee DS, Dollard GJ. Uncertainties in current estimates of emissions of ammonia in the United Kingdom. Environ Pollut 1994;86:267–77.
- Lockyer DR, Whitehead DC. Volatilization of ammonia from cattle urine applied to grassland. Soil Biol Biochem 1990;22:1137–42.
- Lockyer DR, Pain BF, Klarenbeek JV. Ammonia emissions from cattle, pig and poultry wastes applied to pasture. Environ. Pollut. 1989;56:
- Menzi H, Katz PE, Fahrni M, Neftel A, Frick R. A simple empirical model based on regression analysis to estimate ammonia emissions after manure application. Atmos Environ 1998;32:301–7.
- Milford C, Theobald MR, Nemitz E, Sutton MA. Long-term measurements of land-atmosphere exchange of ammonia over grassland, abstract. Sixth International Conference on Air-Surface Exchange of Gases and Particles, Edinburgh, Scotland; 2000 July 3-7. p. 21.
- Moal JF, Martinez J, Guiziou F, Coste CM. Ammonia volatilization following surface-applied pig and cattle slurry in France. J Agric Sci 1995; 125:245-52.
- National Interagency Fire Center. http://www.nifc.gov. Accessed fall 2000. Patni NK, Jui PY. Nitrogen concentration variability in dairy-cattle slurry stored in farm tanks. Trans Am Soc Agric Eng 1991;34:609–15.
- Pollet I, Christiaens J, Van Langenhove H. Determination of the ammonia emission from cubicle houses for dairy cows based on a mass balance. J Agric Eng Res 1998;71:239–48.
- Quality Deer Management Association. http://www.qdma.com. Accessed Fall 2000.
- Rocky Mountain Elk Foundation. http://www.rmef.org/. Status of Elk in North America 1975–1995.
- Roe SM, Strait RP, Niederreiter ML. Methods for improving national ammonia emission estimates. Technical Memorandum, EH Pechan and Associates, Rancho Cordova, California; 1998 May. p. 1–19.
- Seinfeld JH, Pandis SN. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. New York, NY: Wiley; 1998. p. 523-39.
- Sommer SG, Olesen JE. Waste management: effects of dry matter content and temperature on ammonia loss from surface-applied cattle slurry. J Environ Qual 1991;20:679–83.
- Sommer SG, Olesen JE, Christensen BT. Effects of temperature, wind speed and air humidity on ammonia volatilization from surface applied cattle slurry. J Agric Sci 1991;117:91–100.

- Sonoma Technology, Technical Support Study 15: Evaluation and Improvement of Methods for Determining Ammonia Emissions in the San Joaquin Valley. Prepared for the California Air Resources Board, Sacramento, California January; 1998. p. 8-1 to 8-4.
- Strader R, Anderson NJ, Davidson CI. User Guide CMU NH3 Inventory, Version 1.2, http://www.envinst.cmu.edu/nh3/cmunh3userguide.pdf. Accessed April 2001.
- Thompson RB, Ryden JC, Lockyer DR. Fate of nitrogen in cattle slurry following surface application or injection to grassland. J Soil Sci 1987;38:689–700.
- US Census Bureau. United States Census, 1990. http://www.census.gov. Accessed Summer 2000.
- US Census Bureau. United States Census, 1997. http://www.census.gov. Accessed Summer 2000.
- US Department of Agriculture. 1997 Census of Agriculture, United States Summary and State Data, March 1999; vol VII: 28-37, 377-435.
- US Environmental Protection Agency. Toxic Release Inventory, 1995. http://www.epa.gov/tri/. US Environmental Protection Agency. Accessed Summer 2000
- US Environmental Protection Agency (EPA). Air Quality Criteria for Particulate Matter. EPA 600/P-95/001aF, Office of Air Quality Planning and Standards, US Environmental Protection Agency, Research Triangle Park, NC; 1996. p. 5-1 to 5-110.
- US Environmental Protection Agency. National Air Pollutant Emission Trends, 1900–1996. EPA-454/R-98-008, Office of Air Quality Planning and Standards, US Environmental Protection Agency, Research Triangle Park, NC; 1998. p. 4-2.
- Walker JT, Aneja VP, Dickey DA. Atmospheric transport and wet deposition of ammonium in North Carolina. Atmos Environ 2000;34: 3407–18.
- Warneck P. Chemistry of the Natural Atmosphere. New York: Academic Press; 1988. p. 429–515.
- Whitehead DC. Atmospheric ammonia in relation to grassland agriculture and livestock production. Soil Use Manage 1990;6:63-5.

Further reading. List of references for ammonia emission factor studies

- 1. Dairy cattle ammonia emissions from animals in the stable
- 2. Dairy cattle ammonia emissions from waste storage
- 3. Dairy cattle ammonia emissions from spreading of waste
- 4. Dairy cattle ammonia emissions from animals while grazing
- 5. Hog and pig ammonia emissions
- 6. Poultry ammonia emissions

The numbers at the end of each reference refer to the above sources of ammonia emissions (1-6). References found in the main list of references are cited as in the body of the paper.

Aneja VP, Bunton B, Walker JT, Malik BP. Measurement and analysis of atmospheric ammonia emissions from aerobic lagoons. Atmos Environ 2001;35:1949–58. (5)

Aneja et al. 2000. (5)

ApSimon HM, Kruse M, Bell JNB. Ammonia emissions and their role in acid deposition. Atmos Environ 1987;21:1939–46. (5, 6)

Asman 1992. (1, 3, 4, 5, 6)

Asman WAH. Ammonia emission in Europe: Updated emission and emission variations. Report 228471008, RIMV, Bilthoven, The Netherlands; 1992. (4)

Ball PR, Keeney DR, Theobald PW, Nes P. Nitrogen balance in urineaffected areas of a New Zealand pasture. Agron J 1979;71:309-14. (4)

Barton DL, Beauchamp EG. Nitrogen losses from swine housings. Agric Wastes 1986;15:59-74. (5)

Battye et al. 1994. (3, 5, 6)

Bouldin DR, Klausner SD, Reid WS. Use of nitrogen from manure.

In: Hauck RD, editor. Nitrogen in Crop Production. Amer Soc Agron; 1984. p. 221-45. (2, 6)

Bouwman et al. 1997. (3, 4, 5, 6)

Bouwman, Van der Hoek 1997. (5, 6)

Buijsman E, Maas HFM, Asman WAH. Anthropogenic NH₃ emissions in Europe, Atmos Environ 1987;21:1099–22. (2, 3, 4, 5, 6)

Bulley NR, Holbek N. Nitrogen mass balances for dairy farms from feed to field. Can Agric Eng 1982;24:19–23. (2, 3)

Burnhill P, Chalmers A, Fairgrieve J. The British survey of fertiliser practice—fertiliser use on farm crops, 1993. Her Majesty's Stationary Office, London, UK; 1994. (4)

Bussink DW. Relationships between ammonia volatilization and nitrogen fertilizer application rate, intake and excretion of herbage nitrogen by cattle on grazed swards. Fert Res 1994;38:111–21. (4)

Carran RD, Ball PR, Theobald PW, Collins ME. Soil nitrogen balances in urine-affected areas under two moisture regimes in Southland. New Zealand J Exp Agric 1982;10:377–81. (4)

Cass et al. 1982. (5, 6)

Commission of the European Communities. Information on Agriculture No. 47, Luxemburg; 1978. (3, 5)

De Bode MJC. Odour and ammonia emissions from manure storage. Odour and Ammonia Emissions from Livestock Farming. Elsevier, London; 1991. p. 59–66. (5)

Demmers TGM, Burgess LR, Short JL, Phillips VR, Clark JA, Wathes CM. First experiences with methods to measure ammonia emissions from naturally ventilated cattle buildings in the UK. Atmos Environ 1998; 32:285–93. (1)

Dou Z, Kohn RA, Ferguson JD, Boston RC, Newbold JD. Managing nitrogen on dairy farms: An integrated approach. I. Model description. J Dairy Sci 1996;79:2071–80. (2)

ECETOC. Ammonia Emissions to Air in Western Europe. Technical Report No. 62. Brussels, Belgium; July. 1994. p. 1–190. (1, 2, 3, 4, 5)

Pennsylvania Department of Environmental Resources. Field application of manure: A Supplement to Manure Management for Environmental Protection. Document, F.A., Pennsylvania, Department of Environmental Resources, Harrisburg, PA; 1986. p. 1–13. (2)

Genermont et al. 1998. (3)

Giddens J, Rao AM. Effect of incubation and contact with soil on microbial and nitrogen changes in poultry manure. J Environ Qual 1975;4: 275-8 (6)

Gilbertson CB, Van Dyne DL, Clanton CJ, White RK. Estimating the quantity and constituents in livestock and poultry manure residue as reflected by management systems. Amer Soc Agric Eng 1979;22: 602–11. (2, 5, 6)

Harris BD, Thompson Jr EL. Evaluation of ammonia emissions from swine operations in North Carolina. Proceedings of Conference on Emission Inventory: Living in a Global Environment, VIP-88. Air and Waste Management Association, Pittsburgh, PA; 1998. p. 420-9.

Harriss RC, Michaels JT. Sources of atmospheric ammonia. Proc. 2nd Symp, Non -Urban Troposphere. Amer Met Soc, Williamsburg, Canada; 1982. p. 33–5. (5, 6)

Jarvis SC. Nitrogen cycling and losses from dairy farms. Soil Use Manage 1993;9:99-105. (1, 3, 4)

Jarvis, Pain 1990 (3, 5, 6)

Jarvis SC, Hatch DJ, Lockyer DR. Ammonia fluxes from grazed grassland: annual losses from cattle production systems and their relation to nitrogen inputs. J Agric Sci 1989;113:99–108. (4)

Kruse M, ApSimon HM, Bell JNB. Validity and uncertainty in the calculation of an emission inventory for ammonia arising from agriculture in Great Britain. Environ Pollut 1989;56:237–57. (5, 6)

Lee, Dollard 1994. (5, 6)

Lockyer David, R. A system for the measurement in the field of losses of ammonia through volatilization. J Sci Food Agric 1984; 35:837-48. (4)

Lockyer, Whitehead 1990. (3)

Lockyer et al. 1989. (3, 5, 6,)

Menzi et al. 1998. (3)

- Misenheimer DC, Warn TE, Zelmanowitz S. Ammonia emission factors for the NAPAP emission inventory. EPA/600/7-87/001. US Environmental Protection Agency, Washington, DC, USA; 1987. p. 1–4.
- Moal et al. 1995. (5)
- Moller D, Schieferdecker H. Ammonia emission and deposition of NH_x in the GDR. Atmos Environ 1989;23:1187–93. (5, 6)
- Muck RE, Steenhuis TS. Nitrogen losses from manure storages. Agric Wastes 1982;4:41-54. (2)
- Muck RE, Richards BK. Losses of manurial nitrogen in free-stall barns. Agric Wastes 1983;7:65-79. (1)
- Oosthoek J, Kroadsma W, Hoelesma P. Ammonia emission from dairy and pig housing systems. In: Nielsen V, Voorburg J, L'Hermite P, editors. Odour and Ammonia Emissions from Livestock Farming, V. Elsevier, New York, NY 11001; p. 31–41. (1)
- Pain BF, Rees YJ, Lockyer DR. Odour and ammonia emission following the application of pig and cattle slurry to land. In: Nielsen VC, Voorburg JH, L'Hermite P, editors. Volatile emissions from livestock farming and sewage operations. Elsevier, London; 1988. p. 2–11. (3, 5)
- Pain BF, Thompson RB, Rees YJ, Skinner JH. Reducing gaseous losses of nitrogen from cattle slurry applied to grassland by the use of additives. J Sci Food Agric 1990;50:141–53. (3)
- Pain BF, Thompson RB. Ammonia volatilization from livestock slurries applied to land. In: Hansen JA, Henrikson K, editors. Nitrogen in organic wastes applied to soils. Academic Press, London; 1989. p. 202-12. (5, 6)
- Pain BF, Van Der Weerden TJ, Chambers BJ, Phillips VR, Jarvis SC. A new inventory for ammonia emissions from U.K. agriculture. Atmos Environ 1998;32:309-13. (1, 2, 3, 4)
- Petersen SO, Sommer SG, Aaes O, Soegaard K. Ammonia losses from urine and dung of grazing cattle: Effect of N intake. Atmos Environ 1998;32:295–300. (4)
- Ryden JC, Whitehead DC, Lockyer DR, Thompson RB, Skinner JH, Garwood EA. Ammonia emission from grassland and livestock production systems in the UK. Environ Pollut 1987;48:173–84. (3)
- Safley Jr, M, Westerman PW, Barker JC. Fresh dairy manure characteristics and barnlot nutrient losses. Proc 5th Internat Symp on Agric Wastes.

- American Society of Agricultural Engineers, St. Joseph, MI; 1985. p. 191. (1, 2)
- Schlesinger WH, Hartley AE. A global budget for atmospheric NH₃. Biogeochem 1992;15:191–211. (5, 6)
- Sherlock RR, Goh KM. Dynamics of ammonia volatilization from simulated urine patches and aqueous urea applied to pasture: 1. Field experiments. Fert Res 1984;5:181–95. (4)
- Sobel AT. High rise system of manure management. AWM-76-01, Department of Agricultural Engineering, Cornell University, Ithaca, NY; 1976. (6)
- Sommer, Olesen 1991. (3)
- Sutton MA, Place CJ, Eager M, Fowler D, Smith RI. Assessment of the magnitude of ammonia emissions in the United Kingdom. Atmos Environ 1995;29:1393–411. (1, 2, 3, 4)
- Thompson et al. 1987 (3)

Whitehead 1990. (4)

- Van der Hoek KW. Estimating ammonia emission factors in Europe: Summary of the work of the UNECE ammonia expert panel. Atmos Environ 1998;32:315–6. (1, 2, 3, 4, 5, 6)
- Vanderholm DH. Nutrient losses from livestock waste during storage, treatment, and handling. Proc. 3rd Internat Symp on Livestock Wastes. American Society of Agricultural Engineers, England; 1975. p. 282-5.
- van't Ooster A. Using natural ventilation theory and dynamic heat balance modeling for real time prediction of ventilation rates in naturally ventilated livestock houses. Report, N., 94-C-026, XII World Congress on Agricultural Engineering, Milan, Italy 1994, CIGR, Merelbeke; 1994. p. 1–12. (1)
- Vertregt N, Rutgers B. Ammonia volatilization from urine patches in grassland. In: Nielsen VC, Voorburg HH, L'Hermite P, editors. Volatile Emissions from Livestock Farming and Sewage Operations. Elsevier, London; 1987. p. 85–91. (4)
- Warn TE, Zelmanowitz S, Saeger M. Development and Selection of Ammonia Emission Factors for the 1985 NAPAP Emissions Inventory. Office of Research and Development, US Environmental Protection Agency, Research Triangle Park, NC; 1990. p. 11–33. (4, 5)